A comprehensive review of value-added CO2 sequestration in subsurface saline aquifers

A Comprehensive Review of Value-Added CO_2 Sequestration in Subsurface Saline Aquifers

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Abstract

This paper comprehensively reviews CO_2 sequestration process in saline aquifers. The storage mechanisms including structural, residual, solubility, and mineral trappings are assessed along with a discussion of their relative contributions, and their key parameters and optimisations. In view of storage security and capacity, effects of rock and fluid properties and reservoir conditions together with injection strategies are discussed. Furthermore, CO_2 storage site selection is investigated followed by an evaluation of the different measurement, monitoring and verification methods to mitigate the risk of leakage. Field examples with key learnings are also presented to help engineers with sustainable development of storage projects.

Keywords: CO_2 sequestration, Global warming, Saline aquifers, Geological storage, Trapping mechanisms, Leakage monitoring

1. Introduction

Emissions of anthropogenic greenhouse gases (GHG) is proved to be the primary and main cause of the climate change and the subsequent environmental problems (Intergovernmental Panel on Climate Change, IPCC, 2007). Greenhouse gases such as carbon dioxide (CO_2), nitrous oxide (N_2O), methane (CH_4), perfluorocarbons (PFCs), sulphur hexafluoride (SF6) and hydrofluorocarbons (HFCs) contribute to the well-known greenhouse effect. Among all the GHG, carbon dioxide stands out as the most important GHG, not because it is the most potent GHG but rather due to its excessive amount in the atmosphere compared to others. Since the industrial revolution began, CO_2 emission due to human activities and high usage of fossil-fuel based energy

sources have increased significantly that has caused serious climate problems (Goodman et al., 2011; Benson et al., 2012; Blondes et al., 2013; Alcalde et al., 2018). The atmospheric concentration of carbon dioxide as of 2007 was at about 385 ppm, growing steadily at 2 ppm per year compared to that of nitrous oxide (325 ppb) and methane (1780 ppb) (IPCC, 2007). However, the new record as of January 2019 shows that it has increased to 409.92 ppm (NOAA 2019). Different sources of CO_2 emissions and their respective contributions from the year 1980 that is forecasted until 2050, have been illustrated in Figure 1.

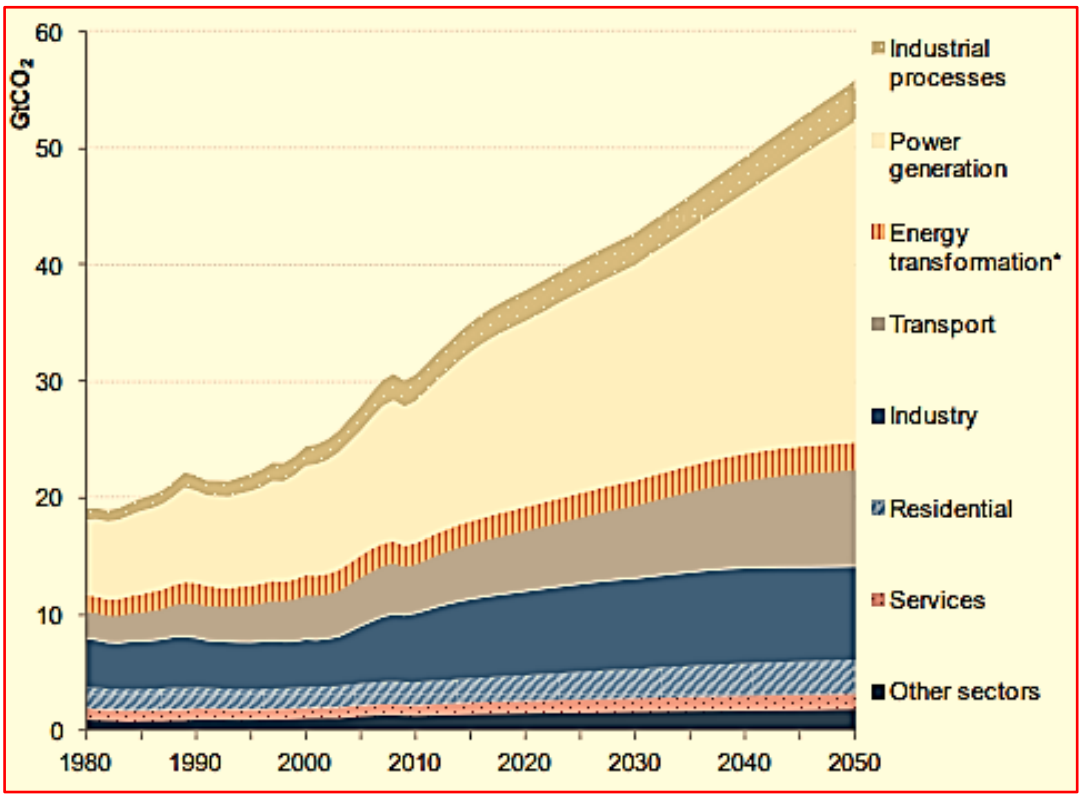


Figure 1: Global CO_2 emissions generated by different sources from (1980-2050) (Marchal et al., 2011)

In order to control the increasing emission and concentration of CO_2 in the atmosphere and mitigate climate change problems, new technologies and approaches need to be employed. Generally, one can cluster the efforts in mitigating climate change into three main approaches, namely, shifting the energy mix to alternative less carbon intensive fuels, energy efficiency improvement and carbon capture and storage (CCS) (Folger, 2017; Gibbins and Chalmers, 2008). It is a fact that, in the near term, a complete shift away from using petroleum-based products is an

almost impossible goal and that energy efficiency alone is not enough to mitigate the rise in emissions. Therefore, CCS that includes CO_2 capture followed by its sequestration in a geological formation is the most promising method to combat global warming which has also been found to be economically feasible (Tcvetkov et al., 2019; Aminu et al., 2017; Liu et al., 2017; Dai et al., 2016; Young-Lorenz 2013; Heddle et al., 2003; Balat and Oz, 2007; Allinson et al., 2003). These geological formations include: (a) deep saline aquifers, (b) depleted oil and gas reservoir, (c) oil and gas reservoirs under CO_2 enhanced oil recovery (CO_2-EOR and Storage), (d) deep un-mineable coal seams, (e) coal bed methane and (f) shale formations during enhanced gas recovery (Heddle at al., 2003; Chadwick et al., 2008; Godec et al., 2011; Blondes et al., 2013, Dai et al., 2014; Foroozesh and Moghaddam, 2015; Dai et al., 2016; Nguyen et al., 2018; Jia et al., 2018; Rezk et al. 2019; Rezk and Foroozesh, 2019; Jiang et al., 2019; Shi and Durucan, 2005). However, selection of an appropriate storage site is crucial and requires suitable strategies depending upon basin suitability, site screening, ranking and characterization (Aminu et al., 2017; Buscheck et al., 2012).

Compared to the rates of terrestrial carbon uptake, only a small amount of CO_2 could be stored into the geological formations per year with 220 million tonnes of man-made CO_2 stored in subsurface formations as of 2017 (Global CCS Institute, 2017). Therefore, sequestration of CO_2 at higher rate are needed to take benefit of the huge availability, capacity and security of such geological formations (Kearns et al., 2017; Bachu et al., 2014).

CO_2 sequestration in subsurface saline geological formations (or aquifers) is considered as the most feasible technology because most of the saline formations in the world are located within the sedimentary basins which are probably highly porous as well as permeable thus have largest storage capacity compared to other geological formations. Moreover, large pores and high permeability of such geological formations make them to require a smaller number of injection wells and also lead to an easier pressure dissipation (Shukla et al., 2010; Aminu et al., 2017). It has been estimated that, the saline aquifers have a storage capacity potential between 400 to 10^4 Gt CO_2 (Bert et al., 2005). Deep saline formations which is widely distributed in U.S. contribute a huge storage capacity resources (approximately 900 Gt–3400 Gt) but still the understanding of effectiveness of trapping mechanisms are unclear at these sites. To provide greater context, Table 1 illustrates the estimated storage capacities for CO_2 available in the United States of America and

North America as a whole for multiple categories of potential storage sites (Spellman, 2014; Shi and Durucan, 2005).

Table 1. Estimated storage capacities in different storage sites in United States and North America (Source: U.S. Department of Energy, National Energy Technology Laboratory, Carbon Storage Atlas, 5th ed., August 20, 2015).

Storage Option	Estimated Capacity (Gt CO_2)	Storage Integrity
Oil and natural gas reservoirs	186-232	High
Unmineable Coal	54-113	Medium
Saline formations	2379-21,633	Highest
Total	2,618 -21,978	

Apart from the huge potential, nowadays saline aquifers are used to recover brine and water by injecting emitted CO_2 from coal industries not only to meet the climate requirement but also to enhance the water security in China (Li et al., 2014 & 2015). This process leads to have huge and secure storage by controlling the pressure as well as to have produced water that can further be used in industries, agricultures and also for home usage after suitable treatments.

The efficiency of the geologic sequestration process mainly depends on the effectiveness of various CO_2 trapping mechanisms. In the case of CO_2 sequestration in saline aquifers, once CO_2 is injected underground into the aquifer, it starts moving up to the top of the formation (as it has a lower density as that of the brine) until it stops and being trapped underneath an impermeable barrier (cap-rock). This physical trapping mechanism, termed structural trapping, can retain CO_2 for long time periods and is influenced by the volume of the trap and the caprock integrity. Some of the injected CO_2 will trap in the pores by capillary forces termed residual trapping. Additionally, part of the injected CO_2 is trapped by dissolving in ground brine known as solubility trapping. Reaction between CO_2 and surrounding rock/minerals also results in trapping which is known as mineral trapping (Rochelle et al., 2004; Farajzadeh et al., 2009). All these trapping mechanisms and the processes change dynamically during and after the injection period of CO_2. The security of these trapping mechanisms and further development to evaluate the potential leakage of sequestered CO_2 back to the surface are the key challenges among the scientists and researchers. Furthermore, economics and environmental risks are essential to be considered for any geologic carbon sequestration project (Li and Liu, 2016; Dean and Tucker, 2017; Castaneda-Herrera et al., 2018). Storage site location and complexity affect the infrastructure costs while depth of formation, rock properties, number of wells and the location (onshore or offshore) are the key factors which

impact the storage cost (Solomon, 2007). Seismic disruption, land surface distortion and contamination of potable water supplies are the key environmental risks that further affect the ecosystems and human health adversely (Cai et al., 2013).

Although having the huge storage potential and current success, addressing the aforementioned criteria and challenges are now essential to accelerate the deployment of this technique. Herein, the recent advances and prospects of CO_2 sequestration techniques in such geological formations have been comprehensively reviewed by including the most updated studies in the literature. The key objective of this review is to convey the importance of trapping mechanisms and governing parameters which affect the storage efficiency, and how these parameters or mechanisms can further be optimised to enhance the storage security based on the past and recent studies. Site suitability, leakage potential and process monitoring have also been incorporated. This article also highlights the challenges associated with CO_2 sequestration and also discusses how these challenges could be technically addressed. Finally, some forthcoming aspects for improving CO_2 trapping and evolving progress in CO_2 sequestration, to make it more effective and economically viable, are discussed.

2. Mechanisms and governing parameters of CO_2 geo-sequestration in saline aquifers

Deep saline aquifers are among the best candidates for sequestration of CO_2 (at industrial scale) due to their considerable capacity and availability. Moreover, the water in such aquifers has high salinity making them unusable. According to the physiochemical and hydrodynamic conditions of the geological formation, the sequestration processes divided into four trapping mechanisms. The different trapping mechanisms are (Juanes et al., 2006):

1. Structural (stratigraphic or hydrodynamic) trapping where injected CO_2 rises and reaches to underneath of the cap rock which further prevents CO_2 from flowing up to the surface.
2. Residual (capillary) trapping where CO_2 become immobile due to prominent effect of capillary forces and relative permeability effects (multiphase flow effect).
3. Solubility trapping, where CO_2 gets dissolve in the formation brine.
4. Mineral trapping as a result of CO_2 and rock minerals reaction leading to precipitation of solid carbonates which is a kind of permanent storage of CO_2.

Figure 2 summarises the different trapping mechanisms at various stages in the life cycle of a typical CO_2 sequestration project. During the injection period, structural trapping is the main

mechanism which prevents migration of CO_2 back to the surface while, mineral trapping is the most secure but the slowest mechanism to store CO_2 in the form of carbonate.

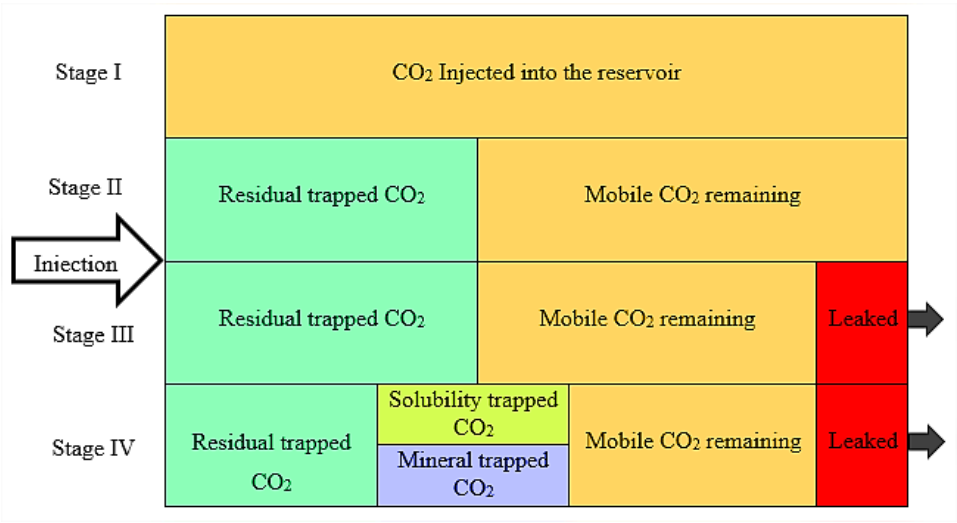


Figure 2: Stages of CO_2 trapping (adopted and modified from Alcalde et al., 2018).

One of the merits of CO_2 during injection into the geological formations is that, CO_2 is in its supercritical state ($scCO_2$) as most of the storage sites exist at depths greater than 800 ft where pressures are likely to be higher than 7.38 MPa (critical point of CO_2). However, CO_2 in its supercritical phase may still undergo change if temperature and pressure change within the reservoir (Rosenbauer et al., 2005). Depending on the reservoir conditions, CO_2 can be stored either in the form of liquid, compressed gas or in its supercritical state. The uncertainty during estimating the net storage capacity, injectivity of CO_2 storage sites and the security can be minimize through a good understanding of governing parameters such as reservoir heterogeneity, depth, permeability, pressure, temperature and their relative effect on different trapping mechanisms. Moreover, the effects of aforesaid parameters regarding to the containment integrity may help in minimizing the risks associated with leakage of CO_2 through different sections in the formation which further prevent any environmental damage. Therefore, the effectiveness of long-term and safe storage of CO_2 in such formations can be improved by optimising the efficiency of active trapping mechanisms. Each trapping mechanisms and their optimal setups are discussed in detail in the following subsections.

2.1 Structural trapping

Structural trapping refers to a time dependent hydrological process where CO_2 is injected into a geological formation below a low-permeable or impermeable caprock in either supercritical or gaseous state and trapped in the formation (Rosenbauer, R.J. and Thomas, B., 2010; Zhang, D. and Song, J., 2014). The density difference between supercritical CO_2 (about 0.6 to 0.7 g/cm^3) and the saline water (1 to 1.05 g/cm^3) in the saline formation results buoyancy force, thus causing the injected CO_2 to uplift to the top part of the aquifer where it is stopped by an impermeable caprock. It should be noted that the vertical and lateral distribution of the injected CO_2 through permeable sedimentary storage rocks underlying the impermeable cap rock, is controlled by the balance between the viscous, capillary, and gravity forces. The types of caprock trapping boundaries are structural traps by anticlines or faults and also stratigraphic traps by an unconformity or a pinch-out (Aminu et al., 2017). The uplifted CO_2 may leak through caprock to the environment affecting the security of stored CO_2. To ensure a secure CO_2 sequestration for a longer period, caprock of the host saline aquifer must have a good sealing capacity as well as the host aquifer must be large enough to store huge amount of CO_2. It is important to mention here that rock wettability in a CO_2-brine system and the interfacial tension have a direct impact on the structural trapping capacity as they control the capillary pressures and relative permeabilities behaviours.

Structural trapping optimisation

Within certain structural trapping scenarios, fluid pressure needs careful management. In the case of some structural/stratigraphic closed systems due to for example fault and pinch-out, while both lateral and vertical movement of the injected CO_2 is contained, the pore pressure can be increased significantly in the vicinity of the injection wells. Bentham and Kirby (2005) have reported that, unconformity of the heterogeneous and structurally compartmentalized reservoirs results in the overburden and geo-mechanical damage of the overlying seals and therefore less suitable for storage of CO_2 compared to large un-faulted or highly permeable geological formations.

As structural trapping is the primary trapping mechanism, it is essential to optimise the responsible parameters to maximise the storage of CO_2. The actual mass of CO_2 which can be trapped by the structural trapping (m_{CO2}) can be estimated by:

$$m_{CO2} = \rho_{CO2} A h \varphi \quad (1)$$

Where h is the height of CO_2 plume trapped under the cap rock, A is the area of the CO_2 plume, ρ_{CO2} is the density of CO_2 and φ is the porosity of the formation. As the mechanism relies on the

very small pores in the pore network of the caprock which results in high CO_2 percolation pressures, thus the storage capacity precisely depends on the net balance of capillary force (P_c) and the buoyancy force (P_b) exerted by CO_2 plume (Iglauer, 2018). The CO_2 plume height permanently immobilized by structural trapping (h) thus can be evaluated by balancing these forces which depends upon the CO_2-brine interfacial tension (σ), brine-CO_2-rock wettability, i.e. contact angle (θ), and the CO_2-brine density difference ($\Delta\rho$) (Eq. 2).

$$h = \frac{2\sigma \cos\theta}{\Delta\rho g r} \quad (2)$$

Where, g is the gravitational constant and r is the average pore radius of the caprock.

Another area of optimisation is the effect of pressure and temperature on brine and CO_2 densities. The brine density decreases monotonically with depth, whereas, the CO_2 density increase with depth because of the high CO_2 compressibility up to a certain depth and becomes independent of depth which results in decreasing nature of $\Delta\rho$ up to a certain depth and becomes constant or less affected by depth. However, at great depth (~15 km), density reversal occurs, i.e. at this depth CO_2 is heavier than water (Wagner and Pruß, 2002; Span and Wagner, 1996). Iglauer (2018) has reported that $\Delta\rho$ decreases from ~1040 to a pseudo minimum ~ 325 (kg/m^3) up to a depth of ~ 1000 m and becomes less affected or somehow constant up to 4000 m. Similar trend has been reported for σ (with depth) in the reported study by Iglauer (2018) which decreases with pressure (below the critical CO_2 pressure of 7.3773 MPa). Apart from the effect of pressure, σ increases with increasing temperature but less dramatic than the effect of pressure. The system wettability or the brine-CO_2-rock contact angle (θ) is not only a function of pressure and temperature; it dramatically increases with an increase in organic content (Arif et al., 2016). Iglauer (2018) has reported the combined effect of pressure, temperature and organic content on θ as a function of depth that explains that θ increases with depth (data are reported up to 2400 m in the study). Additionally, brine composition and rock surface roughness have a direct effect on the wettability of the rock. In summary, the CO_2 column height h can be estimated (based on the above variables in Eq. 2), which decreases with depth. It has been reported in the study that it reaches zero at a depth of ~2400 m and then becomes negative which indicates that, below ~2400 m structural trapping is predicted to fail (because of wettability reversal). However, the actual mass of CO_2 which can be disposed by the structural trapping (m_{CO2}) (in Eq. 1) increases up to a certain depth

of 1300 m (optimal CO_2 storage depth for structural trapping) then decreases and reached zero at ~ 2400 m and enters the negative value.

Injection rate management also plays an important role in optimising structural trapping. It has been found by Raza et al., (2016) that, the amount of free CO_2 (structural trapping) increases with injection rate up to a certain time period and then becomes constant. They have demonstrated the effect of injection rate from 3×10^3 to 3×10^8 sm^3/day and found that monotonic (linear) increase in free CO_2 saturation up to 10 to 20 years depending upon the injection rate and after that decreases due to the rapid and continuous pressure build-up. This decrease in free CO_2 saturation appears at an early stage for higher injection rate.

The geometry of the trapping structure is another key variable when optimising structural trapping. The storage coefficient (E_E) along with volumetric (E_V) and microscopic displacement efficiency (E_d)) is a function of the structure of the closure or the degree of the curvature (flat or dome). It has been found that the more tightly curved formation results in a higher storage coefficient as well as a higher volumetric and microscopic displacement efficiency by concentrating a large amount of CO_2 into a smaller area (Gorecki et al., 2009). Based on the geological variables (E_{geol}) which is 1 for homogeneous formations, Iglauer (2018) reported that reduction in irreducible water saturation (S_{wirr}) leads to an increase in storage efficiency/capacity. Despite of irreducible water saturation (S_{wirr}), storage efficiency strongly depends on the relative permeability characteristics of the system which show that, increase in maximum relative permeability of CO_2 results in decrease in storage efficiency (Okwen et al. 2014). Gorecki et al., (2009) have demonstrated the effect of different governing parameters for the storage coefficient ($E_E = E_{geol} \times E_V \times E_d$) that are tabulated in Table 2.

Table 2. Effect of different governing parameters on storage coefficient, microscopic displacement and volumetric efficiency.

Governing parameters	**E_V**	**E_d**	**E_E**
Structure/degree of curvature/ closure type			
Flat	0.26	0.58	0.15
Quarter Dome	0.28	0.60	0.17
Half Dome	0.29	0.61	0.18
Three quarter Dome	0.38	0.62	0.24
Dome	0.39	0.64	0.25
Depth (m) (constant temperature gradient)			
895	0.14	0.52	0.07
2338	0.23	0.52	0.12

3802		0.28	0.54	0.15
Temperature gradient °C/m (fixed depth and pressure)				
0.020		0.14	0.52	0.07
0.025		0.19	0.54	0.10
0.033		0.15	0.60	0.09
Relative permeability of CO_2 (Kr_{CO2} at S_{wirr}) for Sandstones				
Irreducible water saturation (S_{wirr})	Kr_{CO2} at S_{wirr}			
0.197	0.5265	0.26	0.59	0.16
0.294	0.5446	0.32	0.56	0.18
0.558	0.3319	0.50	0.31	0.15
0.659	0.1156	0.56	0.28	0.16
Vertical to horizontal permeability ratio (effect of anisotropy) (K_v/K_h)				
0.01		0.48	0.35	0.17
0.05		0.32	0.48	0.15
0.1		0.27	0.58	0.16
0.25		0.19	0.64	0.12
0.5		0.19	0.66	0.12
1		0.19	0.67	0.12

"Active storage management" and "active reservoir management" are proved to be an essential tool for optimisation of the storage efficiency (Iglauer, 2018). Le Guénan and Rohmer (2011) analysed that, out of various strategies to control pressure build-up, only "producing at a distant well without stopping injection" leads to an increase in CO_2 storage capacity (or storage efficiency). To reduce the injection rate along with controlled build-up pressure, pressure management by increasing the number of injection wells are one of the techniques to optimise storage efficiency (Bergmo et al. 2011). It could also be achieved by passive brine extraction with simultaneous CO_2 injection. It was found that the CO_2 storage capacity significantly increases by having production wells to produce formation brine during the CO_2 injection (storage) process (Li et al., 2014; Buscheck et al., 2012).

2.2 Residual trapping

When CO_2 is injected into a subsurface formation such as a saline aquifer, the dynamic of two-phase flow of water-CO_2 system would be affected by capillary forces. Capillary pressure effect can cause the CO_2, as a non-wetting phase, to be disconnected/snapped off and residually trapped within the pores (Altman et al. 2014). However, the trapped residual CO_2 which are essentially immobile would be dissolved in the formation fluids by molecular diffusion until an equilibrium concentration is reached. Residual trapping, which is also known as capillary trapping, plays a key contribution in defining the eventual amount of CO_2 migration and distribution within the

formation which, in turn, affects the effectiveness of other trapping mechanisms (Niu et al., 2014). Furthermore, the residual trapping is recognised as a substantial mechanism for storage-security. Holtz (2002) have reported in their 3D reservoir modelling work that, depending upon the porosity and permeability of the formation, capillary trapping ranges could even exceed up to 25% which is generally around 15–25% for a typical storage formation. Hesse et al., (2008) and Ide et al., (2007) reported that, only capillary trapping could achieve immobilization of 100% of the CO_2 in a subsurface plume over-time.

Residual trapping optimisation

In saline aquifers, the pore spaces are highly irregular in shape with having corners (Li et al., 2017). The wetting phase 'brine' (present in the pores of the saline aquifer) would be trapped in pore corners as a residual liquid, and non-wetting phase (injected CO_2) cannot easily drain this residual liquid from the corners during the drainage process (Li et al., 2017; Pini et al., 2012). The CO_2 storage by residual trapping is mainly controlled by the factors affecting the flow of the wetting layer, which are: piston-like (frontal) advance and snap-off processes. In the piston-like (frontal) advance, the water pushes out the CO_2 from the centre of the throats by filling the narrower sections of the pores which have higher capillary pressures. This will end up with reducing the CO_2 residual trapping as the throats and pores will be filled by the water eventually. On the other side, in the snap-off process, the residual water in pores causes swelling of the in-contact rock matrix. This will cause the CO_2 to be trapped in pores by the water that moves to fill the centre of the pore throats. Hence, the frontal advance process reduces the CO_2 residual trapping and the snap-off improves it. Therefore, the combination of these two mechanisms decides about the contribution of the residual trapping to the overall storage process.

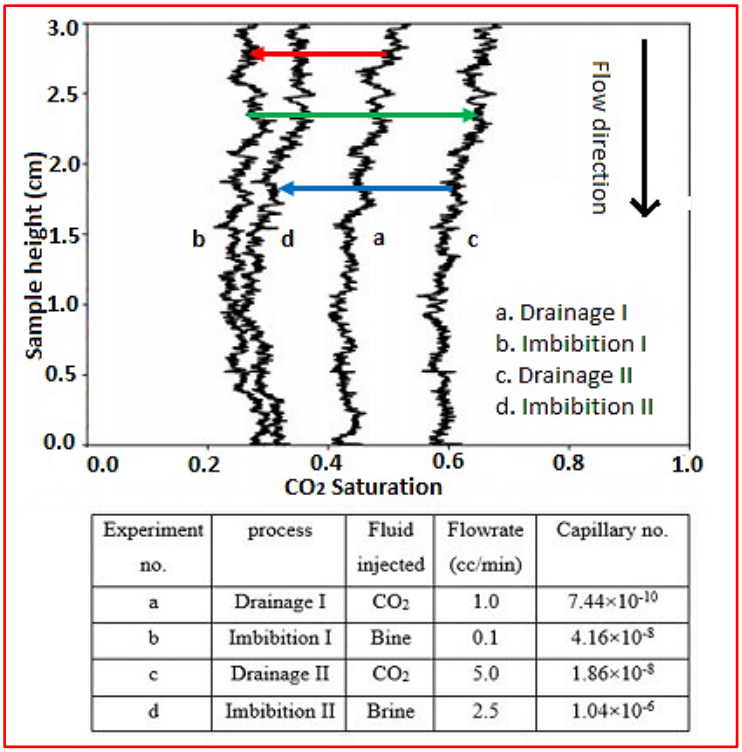
The trapping of wetting phase (brine) or residual water formation significantly depends on the properties of wetting and no-wetting phases as well as rock properties such as contact angle of CO_2-brine-rock system, CO_2-brine IFT (interfacial tension), porosity, permeability and CO_2 injection rate (Li et al., 2017; Li et al., 2015; Wildenschild et al., 2011; Han et al., 2011). A high IFT and contact angle results in high capillary pressure condition which reduces the drainage ability of CO_2, hence lowers the capillary or residual trapping potential (as less water can be displaced to free the space for CO_2 to reside). The IFT and the contact angle are greatly influenced by the brine properties such as total salinity and ion composition as well as pressure and temperature (Li et al., 2017). However, larger pores and throats of the saline aquifers reduces the

capillary pressure (pore radius is inversely proportional to the capillary pressure), and hence decreases the residual water saturation leading to an increase in CO_2 storage capacity. It has been found that, at lower CO_2 injection rates (for longer time) the formation of residual water reduces, and hence more amount of CO_2 can be stored (Wildenschild et al., 2011).

To optimise residual trapping, multiple governing factors that may affect the efficiency of residual trapping have been identified such as pore network, pore restrictions, pore size/grain size, porosity/permeability, wettability and mineral types, injection rate, saturation, conductance, and co-contaminant.

It has been found that, residual CO_2 trapping increases with an increase in permeability (in both horizontal and vertical directions), porosity, and brine density (Han et al., 2011). Suekane et al., (2011) investigated the capillary trapping mechanism using micro-focused X-ray CT at pore scale by trapping the CO_2 and the exploring the effect of grain size/pore size on residual gas saturation in a glass bead packed both at atmospheric conditions and reservoir conditions (elevated temperature and pressure). The residual gas saturations were found higher at lab conditions (20%) than reservoir conditions (16%). They also have found that there is a monotonic increase in residual gas saturation for both atmospheric and reservoir conditions with decrease in glass bead diameter because of dominating behaviour of capillary over viscous effect.

Residual CO_2 saturation as a result of residual trapping mechanism can be controlled by injection history and flow rate. Li et al., (2015) have demonstrated the effect of injection history and flow rate on trapping efficiency by capillary trapping in a Berea sandstone through sequential drainage (brine displacement by CO_2) and imbibition (CO_2 displacement by brine) cycles for CO_2-brine system. It has been found that, an increase in flow rate results in an increase in trapped amount of CO_2 for both the drainage (around 20% increase) and imbibition (approximate 5% increase) as shown in Figure 3.



Experiment no.	process	Fluid injected	Flowrate (cc/min)	Capillary no.
a	Drainage I	CO_2	1.0	7.44×10^{-10}
b	Imbibition I	Bine	0.1	4.16×10^{-8}
c	Drainage II	CO_2	5.0	1.86×10^{-8}
d	Imbibition II	Brine	2.5	1.04×10^{-6}

Figure 3: CO_2 saturation profiles of two consecutive drainage- imbibition cycles on a Berea sandstone sample (adopted and modified from Li et al., 2015)

Altundas et al., (2011) showed that the migration of CO_2 plume can be significantly retarded by considering capillary pressure (Pc) hysteresis. In their study, the authors tried to quantify the effects of relative permeability and capillary pressure hysteresis during post-injection of CO_2 using numerical simulation. They observed the occurrence of residual trapped zone as a result of relative permeability hysteresis, while, capillary pressure hysteresis could often counterbalance the buoyancy force reducing the upward migration of the CO_2. It has also been shown that, the plume migration in the radial direction was restricted because of insufficient CO_2 pressure build-up at the leading edge. On considering the effect of Pc hysteresis, it has been realized that the CO_2 saturation was much higher at the trailing edge and much lower at the leading edge of the plume. However, in the absence of the Pc hysteresis thicker migration was occurred up to an additional 200m distance (in their study).

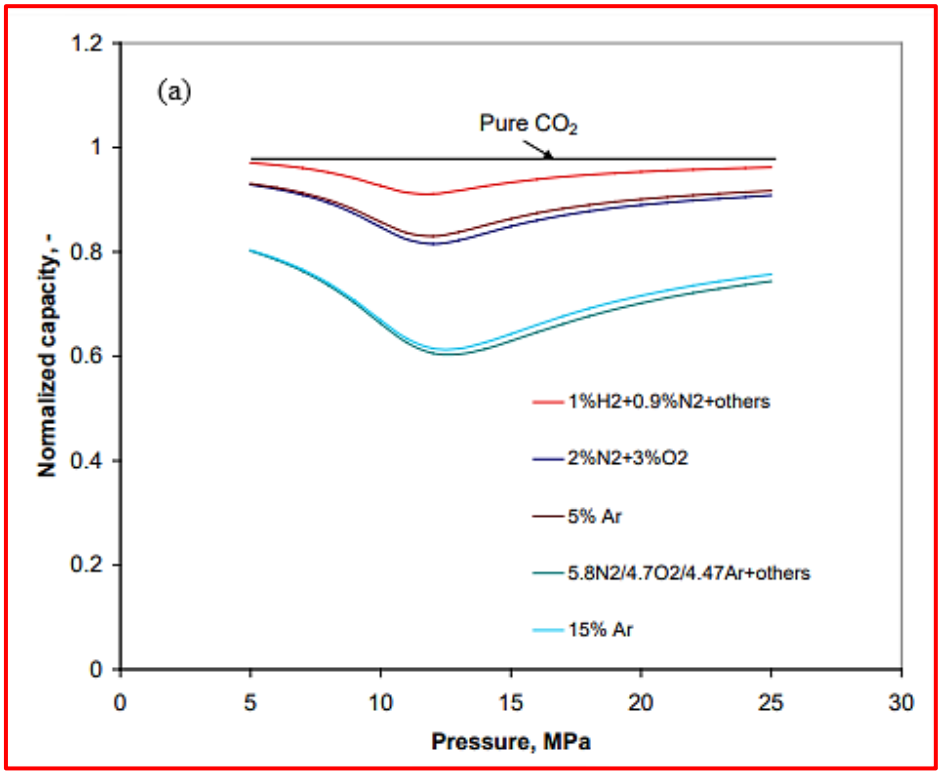
The residual trapping of CO_2 was shown to be significant by Sifuentes et al., (2009) who conducted a series of numerical compositional study to access key contributors to CO_2 storage. In their study, a significant decrease in the amount of mobile CO_2 after injection period was observed due to the relative permeability hysteresis which confirms the importance of the imbibition process

on the residual trapping mechanism. The authors then concluded residual gas saturation of CO_2, S_{gr}, has the largest contribution to the amount of CO_2 stored and by using WAG-like techniques in injecting CO_2 in their study, the amount of mobile CO_2 was reduced from 40% to 20%.

On the numerical modelling front, Kumar et al., (2005) used a compositional simulator to simulate CO_2 storage in order to understand and quantify the importance of residual trapping relative to other competing mechanisms. The injection period was 50 years and the simulation was allowed to continue until 10,000 years to capture the flow of CO_2 saturated brine due to density differences. Although it is known that the solubility of CO_2 into brine phase decreases with an increase in temperature and salinity, the results showed that for a given salinity, the total amount of CO_2 stored in brine increased after 1000 years with increasing temperature. This counter-intuitive result was the consequence of the decrease in density of CO_2 as temperature increases. First, the volume of CO_2 is larger at elevated temperatures, resulting in contact with a larger amount of brine and increases the amount of dissolved CO_2. Second, at higher temperatures, there is higher density dissimilarity between the CO_2 and brine phases that results in rapid movement of the gas and mixing with more brine during upward movement. These two effects led to much CO_2 to be stored in the brine phase at higher temperature (although the solubility of CO_2 in brine decreases). Preliminary results showed that for small values of S_{gr}, nearly half of the CO_2 was still mobile after 1000 years. At larger values, most of the CO_2 was trapped as residual gas after 1000 years. The authors then concluded that the effect of residual gas on CO_2 storage is extremely large and even more significant than solubility trapping and mineralization.

Depending on the source and capturing process of CO_2, presence of different co-contaminants in the CO_2 rich phase would potentially affect the residual trapping efficiency as well as the capacity and injectivity. These contaminants include O_2, N_2, H_2O, SO_X, H_2, CH_4, H_2S, Ar, CO and particulate matters and micro impurities such as HCL, HF, Hg and several heavy metals (Wang et al., 2011). Based on the phase behaviour of pure CO_2 in presence of different impurities, different impurities have distinct effect regardless of their concentrations. It has been found that, non-condensable impurities such as O_2, N_2, and Ar would increase the saturation pressure of liquid CO_2 and decrease the critical temperature. These impurities would also increase the injection pressure and reduce the storage capacity by decreasing not only the volume fraction but also the density of the liquid CO_2 (as shown in Figure 4(a)). These impurities also increase the interfacial tension which results in the decrease in efficiency of residual trapping. Presence of impurities having

higher critical temperature than CO_2, such as SO_2 increases the storage capacity by creating the extra spaces for CO_2 in the pores rather than the chemical effects on the rock surfaces as shown in Figure 4(b). Rasmusson et al., (2018) have discussed the effect of SO_2 on residual trapping with varying salinity and thermodynamic conditions in detail. They have found that, presence of 1 wt.% SO_2 results in increasing of the trapped amount of CO_2 by 3%. However, it should be noted that the presence of SO_2 may cause leakage issue due it its reactivity with caprock.



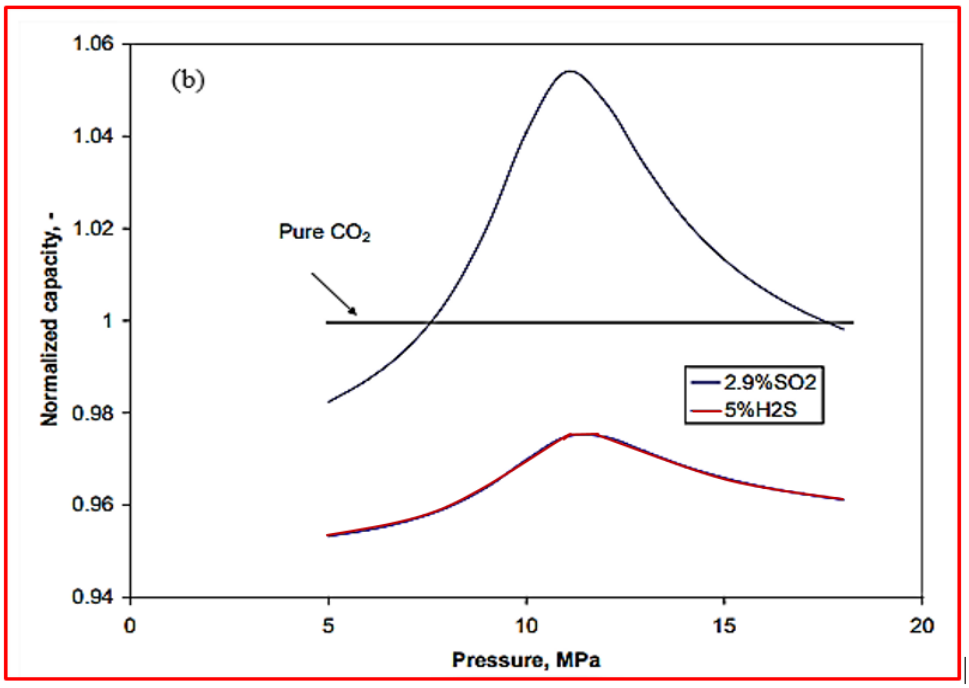


Figure 4: Normalized CO_2 storage capacity in presence of different impurities at 303 K as a function of pressure (adopted from Wang et al., 2011).

Emphasising on the reservoir rock properties data coupled with the reservoir simulation could be the effective tool to estimate the residual trapping efficiency. The percentage of residually trapped CO_2 (R) can be estimated using the ratio of trapped CO_2 saturation (S_t) to maximum CO_2 saturation (S_{max}). Burnside and Naylor (2014) have summarized the value of R for different sandstones, carbonates and shales depending upon the mean porosity, permeability, relative permeability of CO_2 at maximum CO_2 saturation (S_{max}) ($k_r^{CO_2}$), S_{max} and S_t presented in Table 3.

Table 3. Reported experimental values for residual saturation trapping (R).

Lithology	$k_r^{CO_2}$	S_{max}	S_t	**R (%)**	R_{mean} **(%)**
Sandstones (35 samples)	0.06-0.61	0.31-0.85	0.10-0.52	12.8-91.6	61
Carbonates (13 samples)	0.04-0.61	0.14-0.78	0.04-0.41	30.4-74.2	50
Shales (2 samples)	0.19,0.015	0.36,0.39	0.26, 0.35	70.7,88.4	80

It has been believed that, considering the effect of maximum saturation especially from multidirectional core samples may help in predicting the efficiency and heterogeneity of residual trapping. It has also been suggested that cyclic multiphase flow could potentially be used to increase supercritical CO_2 ($scCO_2$) trapping for sequestration applications (Herring et al., 2016). They have estimated that, residual $scCO_2$ saturations of up to 0.50 are achieved after the third D-I cycle, significantly higher than any previous reports. It has also been shown that low vertical permeability and water injection at a larger depth favour residual gas trapping (Nghiem et al., 2009).

2.3 Solubility trapping

Upon injection of CO_2 into the formation, it migrates upward by the effect of density difference, until being trapped by the caprock at the top of the reservoir as it is previously illustrated. Thereafter, the dissolution of CO_2 started at the interface separating the CO_2 plume and brine due to molecular diffusion process. This process is known as the solubility trapping. The CO_2 dissolution in the brine phase causes an increase in the brine density by 0.1% to 1% depending on the reservoir conditions, that results in system instabilities and convective mixing appeared by density-driven natural convection. The convective mixing process results in accelerating the CO_2 dissolution process that can last for long time if only controlled by molecular diffusion (Zhang and Song, 2014).

Solubility trapping optimisation

Leonenko & Keith (2008) carried out a numerical simulation and economics study to justify the performance of brine injection on top of CO_2 gas cap to accelerate dissolution of CO_2. The study was motivated by the results obtained by Hassanzadeh et al. (2007) which revealed that it would need a very long time (500 years) for convection to fully develop in the brine underneath the CO_2 gas cap, resulting in the dissolution of 8% of the CO_2 gas cap. Interestingly, convection appears to be stronger in the case of CO_2 with brine injection versus without brine injection. In the case of brine injection on top of the CO_2 cap, it was observed that the residually trapped CO_2 left below the receding bubble of free CO_2 produced a plume of saturated brine and this plume was much larger as compared to the layer of saturated brine produced in the base case (without brine injection) which further resulted in significant density instability and hence a more rapid start of convection. The author finally concluded that based on numerical simulation results for the case

of brine injection, it is possible to accelerate the dissolution process (71% of CO_2 dissolved within 300 years in their study).

Similarly, Hassanzadeh et al., (2009) also studied the acceleration CO_2 dissolution in saline aquifers using a black-oil model instead by simultaneously injection CO_2 and brine into the aquifer. Thereafter, injection of CO_2 is stopped (after 30 years) while injection and production of brine continues for 200 years. On the base case, it was shown that the injection of brine significantly accelerates the dissolution of CO_2. In fact, without brine injection, only a small amount of CO_2 (< 8%) would be trapped due to the dissolution in formation brine after 200 years. Brine injection, however induced more than 50% dissolution of CO_2. The acceleration is deemed beneficial as it reduces the long-term risk of CO_2 leakage when CO_2 is in a free gas state which my leak through any openings in the cap rock. Sensitivity studies, on aquifer (formation) thickness, tilt angle, formation anisotropy and layering were performed and the results of the sensitivity studies were consistent with the one reported by Leonenko & Keith (2008). However, the above studies on injecting brine in CO_2 storage uses only a single relative permeability curve (drainage) and therefore does not take into account the residual CO_2 trapping during imbibition when water imbibes through the trailing edge of the CO_2 plume. In the case of considering only drainage (residual gas saturation S_{gr}=0), the whole CO_2 plume becomes mobile even at very low saturation at the end of the injection period. The mobile plume migrates further and interacts with the bulk of formation brine. Therefore, it is beneficial for the solubility trapping while, the other consequence is that no CO_2 residual was trapped (Sifuentes et al., 2009)

During CO_2 geo-sequestration, CO_2 solubility increases with increase in pressure but decreases with increase in temperature as well as the salinity and pH (Benson and Cole, 2008). Additionally, cap rock/seal slope or dip angle also influences the solubility efficiency (Meng and Jiang, 2014). Meng and Jiang have investigated the effect of cap rock slope using both 2-D and 3-D simulation based on mass transfer rate including dissolution rate. The results showed that, for higher inclination angle, number of fingers (instability) has been reduced as well as weakened the interactions of fingers as shown in Figure 5. It has been observed that the increase in inclination angle smoothens the diffusive boundary layer and causes less numbers of finger fronts which further results in more stable flow. Han et al., (2011) have inclusively reported that, directional permeability is more significant than porosity for dissolution rate during solubility trapping of CO_2.

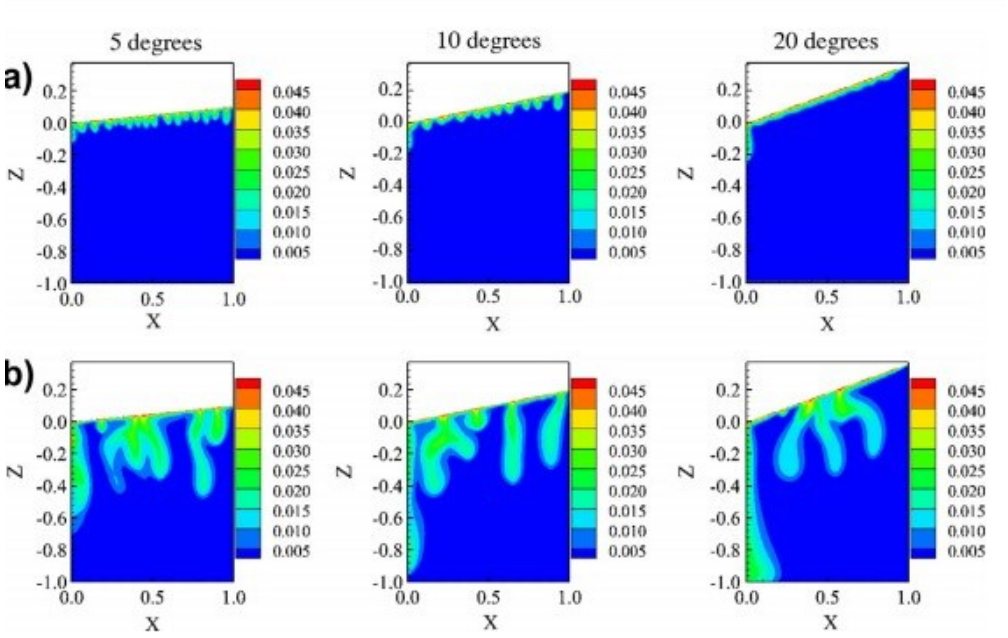


Figure 5: Distribution of the dissolved CO_2 at different time instants: (a) t = 2E5 s and (b) t = 1E6 s for 5°, 10° and 20° of the 2D inclined cases (adopted from Meng and Jiang, 2014).

It was found formation properties (mainly the permeability) control the CO_2 dissolution process that can be further evaluated using dimensionless Rayleigh number (Ra). It is reported that when Rayleigh number is higher than 40, natural convection takes place in the porous media (Lapwood, 1948). In other words, the value of Ra controls the system stability in terms of having natural convection and consequently a higher CO_2 dissolution. It is found that, at high Rayleigh number values, the natural convection will have prominent effect on mass transfer and CO_2 storage. Apart from the effect of permeability, the heterogeneity of geological formation has also a significant effect on the amount of trapped CO_2. Farajzadeh et al., (2011) investigated the effect of heterogeneity on CO_2 solubility. Their study revealed that, depending upon the system heterogeneity three distinct flow regimes can be appeared viz. dispersive flow, fingering and channelling. Moreover, they have also observed a higher mass transfer rate of CO_2 in brine phase in heterogeneous formations. Furthermore, in a sturdy by Green and Ennis-King (2010), the effect of vertical heterogeneity was examined for CO_2 sequestration into deep saline aquifers using randomly distributed shale barriers throughout the aquifer. Because of these shale barriers, convection mixing and consequently the rate of CO_2 mass transfer in the formation brine was reduced. It has also been evaluated that, introducing a single fracture into an aquifer modelled by

Iding and Blunt (2011) enhanced dissolution of CO_2 to 21.6% which was initially 16.9% in absence of fracture. However, the rate of solute transport is highly dependent on the fracture spacing (Shikaze et al., 1998). Recently, Rezk and Foroozesh (2019) investigated the density-driven natural convection during CO_2 sequestration by solubility trapping in fractured heterogeneous saline aquifers. They studied the convective mixing numerically using a single phase 2-D model. In their model, the formation heterogeneity was introduced by spatial variations of permeability using a random function. Furthermore, various realizations of permeability variations were studied for each case to draw reliable conclusions. By introducing a single fracture in a heterogeneous aquifer, their results showed that higher fracture aperture and permeability, and higher inclination towards vertical direction, improved the CO_2 dissolution process. Additionally, by studying the convective mixing in multiple fractured aquifer systems with different orientations, i.e. vertically and horizontally fractured systems, they found that the matrix permeability has the highest impact on the CO_2 dissolution process. Moreover, it was observed that the fracture spacing highly affects the solute distribution and the amount of CO_2 storage in the horizontally fractured aquifers. Their results also showed that increasing the fracture density in the case of vertically fractured aquifers was favourable for the CO_2 dissolution process. As a general conclusion, it was explained that, fractures can improve the convective mixing in aquifers as the fluid velocities are accelerated by the effect of the high permeability fractures that leads to more dissolution trapping in shorter times.

2.4 Mineral trapping

When injected CO_2 dissolves into the formation water (brine) it forms weak carbonic acid which further reacts with the surrounding minerals or organic materials to form solid carbonate mineral (much like as shellfish use calcium and carbon from seawater to form their shells) and other mineral phase which may be beneficial, helping to chemically contain or "trap" the CO_2 as dissolved species or may be deleterious which can cause the migration of CO_2 through the overburden due to change in porosity and permeability of the formation rock. This mechanism is referred as mineral trapping which can be either rapid or slow depending upon the chemistry of the rock and the formation water, but it could immobilize CO_2 or effectively binds CO_2 to the rock for very long periods (Gunter et al. 1997). However, compared to other trapping mechanism, this process is slower and therefore, the overall impact may not be realized for tens to hundreds of years or longer. Mineral trapping takes place over many years at different rates from days to years to thousands of years, but in general, it results in more secure trapping mechanism for CO_2. These

reactions may occur at different parts of the storage system such as formation water, host rock, caprock and borehole completions. The type of reaction depends upon the structure, mineralogy and hydrogeology of the specific lithologies, formation water chemistry, the mineralogical composition of the host rock, the relative rates of the dominant reactions, in-situ temperature and pressure, groundwater flow rates, and also the chemical stability of the engineered features (well completion) (Rochelle et al., 2004). It should be noted that mineral trapping, due to interaction between fluid and rock, is much slower than interaction between CO_2 and the formation water (dissolution trapping) but it provides a more permanent sink for CO_2 and a secure trapping mechanism for CO_2 in the form of carbonate minerals.

Reaction of CO_2 with host aquifer results in an enhanced trapping by buffering the pH of formation water. This effect has been found more prominent in siliciclastic (sandstone) aquifers since siliciclastic aquifers have more potential for pH buffering, solution of CO_2, and precipitation of net carbonate mineral as compared to the carbonate aquifers (Rochelle et al., 2004). Czernichowski-Lauriol et al., (1996) and Pearce et al., (1996) have reported extensive study on reaction between caprock and CO_2 (both dry supercritical and saturated solution) and found these reactions are less well constrained as compared to reaction between CO_2 and host rocks. Interactions between CO_2 and caprock results in initial dissolution of K-feldspar, dolomite, and possibly sheet silicates which may hamper the caprock retention capability to retain CO_2 which leads further migration of CO_2, whereas anhydrite may be barely affected. In general, geochemical reactions can induce fractures in the caprock that cause leakage of the stored CO_2 or it can also inhibit the leakage though precipitations. For example, carbonate minerals can be dissolved that results in high permeability paths in the formation, which facilitates CO_2 leakage. Furthermore, formation water can be dissolved in the injected supercritical CO_2 at high pressures and temperatures, that causes evaporation of the pore water, and consequently induces what is called "shrinkage fractures". On the other hand, the dissolution process as a result of the geochemical reactions may result in precipitation of secondary minerals such as Ca-Mg-Fe carbonates and dawsonite. The deposition of these secondary minerals can reduce the porosity of caprock which results in improved sealing and CO_2 containment potential.

Rochelle et al., (2004) have also reported the reactions involved among CO_2 and the borehole completions such as cement, steel and other subsurface facilities. The understanding of extent of these reactions is crucial which leads to degradation of borehole infrastructure. This further could

lead to migration of CO_2 to shallower formations or directly to the surface. This issue can be mitigated by installing steel well linings and borehole cementation. It is not a permanent solution; however, it may provide suitable containment over a few tens of years.

Mineral trapping optimisation

In order to enlighten the realistic predictions of potential CO_2 sequestration through mineral trapping, an appropriate understanding of the geochemistry of the host formation is crucial. For example, it has been found that the amount of precipitated calcite due to CO_2 reaction primarily depends on the pH of the brine phase rather than the effect of CO_2 pressure and temperature in carbonate formations (Soong et al., 2004). One of the main limitations of the mineral trapping is the slowest nature of the process among other trapping mechanism which requires enhancement in the chemical reactions at different spot of the formation. Enhancement in such reaction by any artificial means at low energy input is still an ongoing research. Keeping the numerous advantages of the mineral trapping mechanism in mind, selection of suitable site based on the favourable conditions for carbonation (such as geothermal gradient) may enhance the storage efficiency together with a reduction in costs. Different studies on CO_2 mineral trapping due to CO_2-water-rock interaction are summarized in Table 4 for different reservoir rock systems.

Table 4. CO_2-water-rock interaction during CO_2 sequestration into geological formations

Rock type	Scope of study	Mechanism	Outcomes	References
Sandstone	CO_2 sequestration potential in the heat mining process applied to geothermal reservoirs	Dissolution of clay and ankerite minerals followed by precipitation of plagioclase, which results in an increase in concentration of Mg^{2+} and Ca^{2+} in formation brine.	Precipitated minerals reduce the porosity and has an adverse impact on the heat mining rate.	Cui et al., 2017
Carbonate		Dissolution of dolomite followed by the precipitation of calcite and ankerite.	The dissolution of clay and dolomite can surpass the precipitation of silicate and calcite minerals and rise the heat mining rate.	
Basalt	Feasibility of CO_2 sequestration at low temperature considering the key	Stability of precipitated mineral depends upon the	1) Fe played an important role in carbonate rocks at low reaction rate.	Gysi and Stefánsson, 2012

	reactions and chemical mass transfer	acidity and basicity of the system.	2) Rapid formation of carbonates was at elevated CO_2 concentrations (pH <6.5). 3) Predominant effect of injected calcium carbonates was seen at pH >6.5.	
Limestone (38% Calcite)	Effect of high temperature and pressure on reaction rates during sequestration in aquifers	Increase or decrease in porosity is function of sulphate concentration in the brine.	1) Enhanced solubility of CO_2 with having rapid geochemical reactions. 2) The solubility of CO_2 is function of pressure and temperature.	Rosenbauer et al., 2005

3. Security of CO_2 sequestration in geological formations for different trapping mechanisms

For a secure and effective CO_2 sequestration with minimum risk of leakage, it is essential to evaluate any storage site technically and economically to ensure a sustainable storage. Alcalde et al., (2018) presented a numerical program called Storage Security Calculator (SSC) based on the established and measured geological process. It was used to assess retention of CO_2 in the formation together with the surface leakage flux rate, to optimise the knowing and predicted assessment for safe storage for a longer period. Based on their study, they have suggested that monitoring regulated subsurface operations and comprehensive site screening could minimize the risk of leakage. Figure 6 represents the life cycle of CO_2 migration evolving over time through different mechanisms and the contrast among different mechanisms based on the security of CO_2 storage. As CO_2 get dissolved in the formation brine, it increases the density of brine. Therefore, CO_2 associate with brine will start sink slowly (due to high density) and its upward migration becomes restricted. This mechanism is known as solubility trapping as we previously discussed, which has an approximate timescale of thousands of years. As it has been already discussed earlier that the most secure but the slowest mechanism is mineral trapping, considering its approximate timescale varies from thousands to billions of years. As such, it becomes least important during the time scale that we are interested in (as shown in Fig. 6). Nevertheless, the most rapid mechanism to store CO_2 and make it immobile is by residual trapping (or capillary trapping). In terms of storage security, essentially, the longer CO_2 can stay sequestered, the more secure it

becomes as geochemical traps start taking effect. Both the extent and rate of trapping of residual trapping through capillarity action made it integral to storage security as compared to the other trapping mechanisms as shown in Figure 6. However, stratigraphic and structural are the initial primary trapping mechanisms for storing the injected CO_2 gas but less secure among other mechanisms due to the greater chance of leakage of the free CO_2. Once the mineralization begins, the CO_2 will no longer be able to escape the reservoir in any aspects which results in highly secure storage with minimum risk of leakage. As the site gets older, less oversight is necessary to ensure its safety. It should be noted that although different trapping mechanisms have different operating timescales, the net result is a gradual increase in the permanence of the stored CO_2. The timespan of these trapping mechanisms are in order of; $t_{str.}$(structural trapping) < $t_{res.}$(residual trapping) << $t_{sol.}$(solubility trapping) <<< $t_{min.}$(mineral trapping).

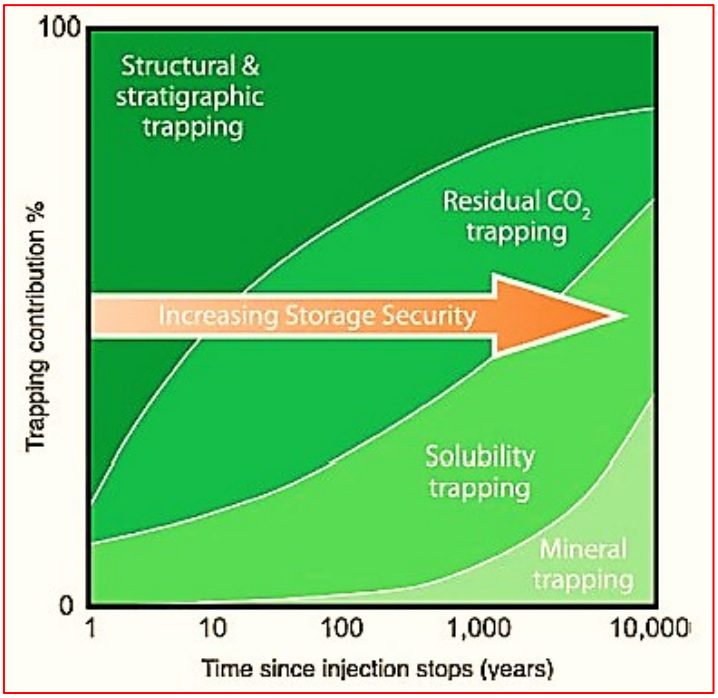


Figure 6: A schematic of trapping contributions of various trapping mechanisms over time and their relative importance based on storage security (adopted from Benson et al., 2012).

4. Site characterization, storage capacity and screening criteria

A CO_2 storage site can be a layered, deeply buried, permeable rock formations but must be overlain by impermeable cap rocks. A CO_2 storage site must also be able to accept and retain injected CO_2 which is an essential component of site assessment before starting any CO_2 injection.

A list of different potential formations for sequestration along with characteristics and governing trapping mechanisms is tabulated in Table 5.

Table 5: List of different potential formations for sequestration of CO_2

Type of Reservoir	**General Characteristics**	**Principal Trapping Mechanism**	**References**
Deep Saline Formations	Sandstone and carbonate (limestone or dolomite) rocks saturated with salty water.	Hydrodynamic, dissolution, and mineralization	Bachu et al., 2014; Balashov et al., 2013
Depleted Oil Reservoirs	During and after EOR operations, CO_2 may be injected or stored either to recover unrecovered oil or direct storage.	Hydrodynamic, dissolution, and mineralization	Godec et al., 2011; Le Gallo et al., 2002
Depleted Natural Gas Reservoirs	Once the natural gas has been produced, it can be used to store CO_2 but specially used for natural gas storage.	Hydrodynamic, dissolution, mineralization	Van der Meer, 2005; Raza et al., 2018
Deep Unmineable Coal Seams	CO_2 can be injected into the coal seams to induce the coal to release its methane leading to CO_2-driven enhanced coalbed methane recovery (ECBM).	Primarily chemical adsorption	Shi, and Durucan, 2005; Vishal, 2017
Deep Saline-Filled Basalt Formations	The mineralization potential of such formation is higher due to the presence of iron and other elements which results in secure storage.	Hydrodynamic, dissolution and mineralization	Goldberg et al., 2008; Matter et al., 2011
Other (Salt Caverns, Organic Shales, etc.)	These reservoirs may provide novel niche CO_2 storage options.	Various mechanisms	Dusseault et al., 2002; Pu et al., 2018

Sequestration of CO_2 requires suitable sites to store large volumes of CO_2 for long periods of time, and this extended timescale of storage leads to demanding regulatory, technical and legal aspects for any storage project, which differ across countries. The purpose of site screening within a large area as the targeted zone, such as the entire of a sedimentary basin, followed by site selection and site characterization, is to evaluate the location of qualified sites ready for permitting. Injectivity, capacity, and effectiveness as well as a satisfactory sealing caprock or confining unit are the most important aspects of storage site characterizations (Friedmann, 2007).

In terms of storage capacity, multiple experimental and simulation studies have been reported to estimate the amount of CO_2 that can be stored in a reservoir along with a brief analysis and then modelling based on the initial and boundaries conditions (Johnson, 2009; Pruess and Birkholzer, 2010; Kearns et al., 2017). The storage capacity can either be estimated using the static approaches

that relay on volumetric and compressibility-based algorithms or dynamic approaches that relays on transient numerical or analytical models (Goodman et al., 2011; Cantucci et al., 2016). However, exact storage capacity estimation in saline aquifers is not that easy due to the dynamics of different physiochemical trapping mechanisms that act simultaneously with different rates in different timescales. Out of all the existing trapping mechanisms, the mineral trapping could make the storage capacity estimation process much complicated due to its complex nature and its poorly-understood timescale and therefore mineral trapping mechanism has not been considered by any storage capacity estimation (Goodman et al., 2011; Aminu et al., 2017). Different methods are available for estimating the CO_2 storage capacity into saline aquifers namely CSLF (Carbon Sequestration Leadership Forum) (Bachu et al., 2007; Goodman et al., 2011), US-DOE (United States Department of Energy) (Goodman et al., 2011), USGS (United States Geological Survey) (Gregersen et al., 1997; Hansen et al., 2005) and pressure-limit method (Zhou et al., 2008; Cantucci et al., 2016; Szulczewski et al., 2012). The CSLF method includes physical trapping (structural and stratigraphic) using volumetric approaches together with residual and solubility trapping using time dependent numerical approaches. US-DOE method only includes physical trapping mechanism using volumetric and compressibility-based approach, while USGS method includes residual trapping based on the boundary conditions with considering the effect of buoyancy. Distinctively, pressure-limit method quantifies the storage capacity based on the maximum allowable pressure sustained by the saline aquifer. However, considering the transient effect of pressure build-up on the pore expansion and brine distribution (due to rock and brine compressibility) facilitates the estimation of the additional CO_2 storage volume (Zhou et al., 2008). The most common methods to evaluate regional storage capacity, used by each of the regional assessments, are variations on the volumetric method based on the available pore space which has been further subjected to various limitations (Vangkilde-Pedersen et al., 2009; Goodman et al., 2011; Blondes et al., 2013; Kearns et al., 2017). Bachu (2010) have extensively reported the methods for evaluating the storage capacity for deep saline aquifers, oil and gas reservoirs as well as for coal beds.

While storage capacity is of paramount importance, from a site screening point of view, there has been increasing emphasis placed on containment efficiency and injectivity. Szulczewksi et al., (2014) noted that a site estimated capacity is constrained from both the pressure build-up as a result of injection of CO_2, which is directly related to the injectivity associated with the formation as well

as the sufficient pore volume to trap CO_2 permanently in a safe manner. The constraint on pressure exists because there is a maximum amount of CO_2 that can be injected before the increase in pressure fractures the formation or causes fault re-activation. The author concluded that generally, an aquifer is often constrained on pressure if the site is shallow and long with low permeability and high porosity. Additionally, injection time is also equally important as short injection time places more weight on the pressure constraint. In other words, a site is pressure constrained if the intended target is to inject a particular volume of CO_2 in a shorter time frame, causing pressure to build-up rapidly. Based on the above-mentioned trapping mechanisms and their optimisation studies, the key parameters have been summarized for each mechanism in Table 6. However, there are still controversies over some of the parameters for their exact potential in individual mechanism and requires further research to come up with more generic concepts.

Table 6. Effect of key parameters on potential of different trapping mechanisms

	Increase trapping potential	**Decrease trapping potential**
Structural trapping	1. Caprock integrity 2. Presence of secondary seal 3. Increasing injection rate 4. Increasing depth (certain) 5. High concentration of $CaCO_3$ into the formation (CO_2 injection rate must be high) 6. Increasing curvature (flat to complete dome)	1. Elevated injection pressure (compared to entry pressure of seal) 2. Leakage 3. Thermal stress in caprock 4. Presence of impurities in injected CO_2 stream
Residual trapping	1. Increasing flowrate 2. Small to intermediate scale heterogeneity 3. Increasing number of wells 4. Horizontal well installation 5. High permeability and porosity 6. High concentration of $CaCO_3$ into the formation (CO_2 injection rate must be high) 7. Increasing brine density 8. High pressure (at lower well count) low pressure (at higher well count)	1. Capillary pressure hysteresis 2. Increasing grain size 3. Intermediate wet reservoir 4. Formation dying effect 5. Present of impurities ($Ar/N_2/O_2$) in injected CO_2 stream
Solubility trapping	1. Increasing flowrate and pressure 2. Small to intermediate scale heterogeneity 3. Co-injection of brine and CO_2 4. Increasing vertical permeability 5. High concentration of $CaCO_3$ into the formation (CO_2 injection rate must be high) 6. Present of non-condensable impurities in injected CO_2 stream	1. Increasing Temperature 2. Increasing salinity 3. Increasing pH 4. Elevated pressure 5. Present of non-condensable impurities ($Ar/N_2/O_2$)

Mineral trapping	1. Increasing pH 2. High ion concentration (Ca^{2+}, Mg^{2+}, Fe^{2+}) 3. High pressure 4. Presence of SO_2 and H_2S in injected stream 5. Availability of nucleation sites 6. Host formation contains significant ferric ion	1. Low Temperature 2. Low pressure 3. Low pH

5. Measurement, monitoring and verification (MMV) components

As a prerequisite for carbon sequestration projects, it is required that the injected CO_2 is confined within the geological formation during and after injection with no unexpected migration or leak occurring, which should be monitored through the project. After site characterization, additional investigations should be performed for capturing the baseline conditions at the site prior to the commencement of CO_2 injection. That is, measurement (modelling and analysis of geology and hydrology of the injection system in beginning), monitoring (tracking the movement of the plume) and verification (verifying that the CO_2 remains confined and there is no leak) programme, abbreviated here to MMV, should be designed and implemented. It is basically concerned with the capability to measure the amount of CO_2 storage at a particular site, to map its spatial migration over time, to develop techniques for the early detection (monitoring) of any leakage and finally to verify that the stored CO_2 is isolated and will not adversely affect the host ecosystem (Dean and Tucker, 2017; Li and Liu, 2016). The MMV plan is specific to the site and tailored to the individual site characterization and risk assessment. Efficient capture and storage require zero tolerance for any leak which further requires a proper MMV even the risk of leakage is small (Themann et al., 2009). Themann et al., (2009) have reported an extensive MMV study for CO_2 storage which covers the entire process during pre-injection, injection and post-injection periods both for the storage site and injection facilities. Dean and Tucker, (2017) have proposed an advanced MMV procedure for Goldeneye field, UK. It enables different advantages while satisfies UK and EU regulations as well as lowering cost and reducing uncertainty and effort when compared with conventional methods. Identifying various potential routes for leakage of CO_2 along with their remediation techniques and the method of early leak detection are the key factor for optimising the effective MMV techniques at right time (Benson and Hepple, 2005; GHG, 2007; Esposito and Benson, 2011). Recently, Castaneda-Herrera et al., (2018) have discussed the different pathways

of potential leakage (engineered and natural) along with their remediation techniques. The remediation of engineered leakage (leakage associated with wells or near the borehole) requires injection of highly viscous materials; whereas, injection of low viscosity materials could be helpful for remediation of natural leakage (through geological formations and structures) since they can occupy a larger area on the caprock. Gelation techniques near the injection point has been gaining potential to mitigate leakage issue; however, it requires accurate monitoring methods (Li et al., 2019). Lots of monitoring techniques have been proposed and implemented in the fields (either pilot or commercial projects) to detect different risks associated with CO_2 sequestration which are tabulated in Table 7.

Table 7: Different risks associated with CO_2 sequestration projects and corresponding Monitoring technologies (Mathieson et al., 2011; Li et al., 2013; Cai et al., 2013; Li and Liu, 2016; Li et al., 2016)

Risk/Parameters	Monitoring technologies
Plume migration	3D Seismic, VSP seismic, Gravity survey, InSAR monitoring, Tiltmeters/GPS, Wellhead/Annulus samples, Tracers,
Underground characteristics	3D Seismic, VSP Seismic, Gravity survey
Caprock integrity	Tiltmeters/GPS, InSAR monitoring, Microseismic, Shallow aquifer wells
Wellbore integrity	CO_2 injection rate and pressure (both bottom-hole and wellhead), Wireline, Pressure of monitoring wells (both bottom-hole and wellhead), Wellhead/Annulus samples,
Pressure development	Tiltmeters/GPS, InSAR monitoring
Potable aquifer contamination	Shallow aquifer wells
Surface seepage	Microbiology, Surface flux/soil Gas
Subsurface characterization	Logging
Isotopic analysis of CO_2 and soil gas composition	Soil gas sampling
Density variation due to the fluid displacement	Time-lapse gravity measurements
Source, Location, and magnitude of seismic events	Passive seismic monitoring
Sonic velocity, Brine salinity and CO_2 saturation	Well logs
CO_2 fluxes between the atmosphere and land surface	Eddy-covariance or flux chambers

A well-structured cost estimation by Benson et al., (2004) and Zahid et al., (2011), Liu (2012) have been reported for different monitoring techniques along with total monitoring cost associated by choices 'basic monitoring package' and 'enhanced monitoring package' for both CO_2-EOR in

hydrocarbon reservoirs and storage in saline formations (based on average cost for low residual gas saturation (LRG) and high residual gas saturation (HRG)) is summarized in Table 8.

Table 8. Cost of monitoring packages (Benson et al., 2004 and Zahid et al., 2011, Liu (2012))

	Monitoring Package Cost, USD						
	Basic				**Enhanced**		
Technologies	**Hydrocarbon reservoir (CO_2-EOR)**	**Saline formation (LRG)**	**Saline formation (HRG)**		**Hydrocarbon reservoir (CO_2-EOR)**	**Saline formation (LRG)**	**Saline formation (HRG)**
	pre-operational monitoring						
Well logs	0	1064250	1064250		0	1064250	1064250
Wellhead pressure	0	55000	55000		0	55000	55000
Formation pressure	0	328000	328000		0	328000	328000
Injection and production rate	0	550000	550000		0	550000	550000
Seismic monitoring	0	3828000	2387000		0	3828000	2387000
Electric and electro-magnetic monitoring	N/A	N/A	N/A		360000	225000	225000
Gravity monitoring	N/A	N/A	N/A		360000	225000	360000
Micro seismicity	475000	475000	475000		475000	475000	475000
Atmospheric CO2 concentration	320000	100000	100000		320000	100000	100000
Surface flux monitoring	N/A	N/A	N/A		700000	700000	700000
Fluid and gas composition	N/A	N/A	N/A		1000000	1000000	1000000
Management (15%)	119250	960038	743888		482250	1282538	1066388
Sub-total	914250	7360288	5703138		3697250	9832788	8310638
	operational monitoring						
Casing Logs	N/A	N/A	N/A		13200000	6000000	6000000
Electric and electro-magnetic monitoring	N/A	N/A	N/A		1440000	936000	936000
Seismic monitoring	15840000	9493000	9493000		15840000	9493000	9493000
Micro seismicity	3675000	3675000	3675000		3675000	3675000	3675000
Gravity monitoring	N/A	N/A	N/A		1440000	936000	936000
Wellhead pressure	1500000	1665000	1665000		1500000	1665000	1665000
Injection and production rate	6450000	3351000	3351000		6450000	3351000	3351000
Atmospheric CO2 concentration	2460000	1800000	1800000		2460000	1800000	1800000
Surface flux monitoring	N/A	N/A	N/A		4800000	4800000	4800000
Fluid and gas composition	N/A	N/A	N/A		570000	570000	570000
Management (15%)	4488840	2997600	2997600		7706340	4983900	4983900
Sub-total	34414440	22981600	22981600		59081940	38209900	38209900
	closure monitoring						
Wellhead pressure	N/A	N/A	N/A		1250000	277500	277500

Surface flux monitoring	N/A	N/A	N/A		3200000	8000000	8000000
Fluid and gas composition	N/A	N/A	N/A		380000	950000	950000
Seismic monitoring	7920000	15983000	11935000		7920000	15983000	11935000
Electric and electro-magnetic monitoring	N/A	N/A	N/A		720000	1519000	1125000
Gravity monitoring	N/A	N/A	N/A		720000	1519000	1125000
Management (15%)	1188000	2397450	1790250		1978500	4237275	3511875
Sub-total	9108000	18380450	13725250		15168500	32485775	26924375
Total cost:	44436690	48722338	42409988		77947690	80528463	73444913
Total Cost at 10% discount	12683389	13697010	12023781		23319093	20927707	19250724
Total CO_2	258E8	2.58E8	2.58E8		2.58E8	2.58E8	2.58E8
Cost/CO_2 tonne	0.172	0.189	0.164		0.295	0.312	0.284
Discount Cost per CO_2 Tonne	0.049	0.053	0.047		0.090	0.081	0.075

6. CO_2 storage field projects

It has been realized and agreed by many experts that CCS is an enormously significant mid-to-long-term solution for mitigating and even nullifying the net GHG emissions before 21st century. In contrast to this the next challenge is to build a powerful industry of similar scale to the oil and gas industry in upcoming few decades while meeting the desired objective stated in different agendas for climate change such as the Kyoto Protocol and Paris Agreement. After the United States; Canada, Europe and China are currently leading the way in the CCS sector. Several pilot and commercial projects for CO_2 sequestration are under way or proposed globally. Different fields such as Snøhvit in the Barents Sea, Gorgon in Australia, Sleipner in the North Sea and In Salah in Algeria are the most relevant and strategic commercial projects that have undertaken large scale CO_2 injection for storage purposes.

Under a CCS project, Statoil, BP and Sonatrack companies have injected stripped CO_2 from natural gas industries into a gas reservoir surrounding to the Salah Gas Field in Algeria. In another case, about 10 Mt CO_2 (at injection rate of 1 Mt CO_2 per year) has been injected into a deep subsea saline formation since 1996 by Statoil at the Sleipner project (Solomon, 2007). Prospects for enhanced oil recovery (EOR) have increased interest in CO_2 storage with simultaneous increase in efficiency of EOR which provides considerable experience and insights for safe, reliable injection and storage of CO_2. A CCS facility has been inaugurated and started at the Boundary Dam coal-fired power station in 2014, in Saskatchewan, Canada. The captured CO_2 from the power plant has been injected into oil wells for enhancing the oil recovery (Folger, 2017). Using such oil and gas field for CO_2-EOR projects results in profitable CCS scheme because it mitigates the GHG as well

as emphasizes on economic value to the projects depending on the oil price. However, this kind of geological resources are very limited which only contribute 10% to carbon reduction targets. In Abu Dhabi, a CCS project captures 800,000 metric tonnes of the emitted CO_2 annually by steel industries, followed by compressing it and sending to an oil field which is 50 kilometres far away for CO_2-EOR (Ustadi et al., 2017). Since 2017, the largest system ever applied to a coal-fired power plant was inaugurated in Texas. The Petra Nova Project has got success for capturing more than 1.5 MtCO_2 emitted annually by the plant (Folger, 2017). Here too, the CO_2 planned to be injected into oil wells which further increase the production around 300 to 15,000 barrels per day.

TOTAL operated a pilot project at the Lacq gas field in south-western France. In its first phase (2010-2013), injected 50,000 metric tonnes into a depleted gas reservoir while, in its second phase (2013-2016), carried out monitoring the reservoir to evaluate long-term environmental impact (TOTAL, 2015).

An innovative, ongoing world-leading and world's largest project (around 80,000 tonnes of CO_2 injected and stored) in south-western Victoria, Australia operated by CO2CRC's Otway research facility is demonstrating that CCS is an ecologically and technically safer way to minimize global GHG emissions. This project also emphasizes on technical evidence on sequestration and monitoring of CO_2 that will stimulus national policy and industries while providing pledge to the community (Ashworth et al. 2010). As another example, Tomakomai pilot project in Japan has been started in order to evaluate the viability of offshore storage aiming to store 100,000 tonnes of CO_2 per year (Tanaka et al., 2014). Several projects carried out in past, ongoing and planned for CO_2 sequestration in geological formation have been tabulated in Table 9.

Table 9: CO_2 sequestration projects in geological formations.

Project	Place	Year started	Scale	Net storage	Storage Type	Lithology	References
Sleipner	Norway	1996	Commercial	20 Mt planned	Aquifer	Sandstone	Rubin and De, 2005
Fenn Big Valley	Canada	1998	Pilot	200 t	CO_2-ECBM	Coal	Rubin and De, 2005
Weyburn	Canada	2000	Commercial	20 Mt planned	CO_2-EOR	Carbonate	Rubin and De, 2005
Qinshui Basin	China	2003	Pilot	150 t	CO_2-ECBM	Coal	Rubin and De, 2005
Salt Creek	USA	2004	Commercial	27 Mt	CO_2-EOR	Sandstone	Rubin and De, 2005
Yubari	Japan	2004	Demo	200 t Planned	CO_2-ECBM	Coal	Rubin and De, 2005
CSEMP	Canada	2005	Pilot	10 kt	CO_2-ECBM	Coal	Rubin and De, 2005
Otway	Australia	2008-2010	Commercial	110.2 ('000 tonnes/yr)	Depleted Gas Reservoir	Sandstone	Young-Lorenz and Lumley, 2013
Snøhvit	Norway	2008	Commercial	700 (kt/yr)	Saline Aquifer	Sandstone	Rai et al. 2008
Ordos storage project	Inner Mongolia (China)	2011	Pilot	0.1 Mt/yr	EOR/Saline aquifer	Sandstone	Luo et al., 2014
HECA	USA	2014	Possible	2496.8 (kt/yr)	EOR		Rai et al. 2008
Tomakomai	Japan	2015	Commercial	250 (kt/yr)	saline aquifers, offshore area of the Tomakomai Port	volcanic and volcaniclastic rocks	Tanaka et al., 2014

7. Economics of CO_2 storage in geological formations

Economic viability of any CO_2 capture and sequestration (CCS) project depends upon the capture cost, transportation cost, storage cost and monitoring cost. Among all, capture is the most expensive component of the CCS process which alone accounts for 70 to 80 % of the total cost while storage, monitoring and verification costs are likely to be the least costly components (Balat and Öz, 2007). In this paper, we have mainly focused on the storage cost followed by monitoring cost. Basically, variation in the storage costs depends upon the geological features of the storage site and whether there is a need to cap any potential leakage points (APH, 2007). Heddle et al., (2003) have broken down the CO_2 storage cost model into three components *viz*., injectivity, pressure change calculation and a set of capital, operating and maintenance cost factors. They have

reported the detailed economics of the whole CCS process chain along with sensitivity analysis without ignoring any major parameters and reviewed various commercial field studies. The cost of geological storage of CO_2 is highly site-specific and dependent on factors such as the depth of the storage formation, permeability of formation rock, the number of wells needed for injection, flow rate and whether the project is onshore or offshore (Balat and Öz, 2007). There may be some revenue granted in the case of CO_2 sequestration in EOR and ECBMR (Enhanced coal bed methane recovery) site depending upon the oil prices, price of coal bed methane (CBM) production and CO_2 recycling factor. However, a net storage cost required without any such revenue in the case of other storage sites such as depleted oil and gas reservoirs, deep saline aquifers, ocean storage (both via pipeline and tanker). It has been estimated that the 'Levelized Annual CO_2 Storage Cost in \$/tonne CO_2 (GHG avoided basis)' is 12.21 for EOR, 5.59 for ECBMR, 4.87 for depleted gas reservoirs, 3.82 for depleted oil reservoirs, 2.93 for aquifers, 5.53 for storage in oceans by pipeline and 17.62 for storage in oceans by tanker which is little higher than others (Heddle et al., 2003). It is expected that, the storage capacity of the site affects the cost of the operation. For example, Vidas et al., (2012) have discussed the relation between available storage capacities to cost for Lower-48 fields (US, geography) using GeoCAT (Geologic Sequestration Cost Analysis Tool). In terms of cost of monitoring, depending upon the strategy and technology used for monitoring, the cost should vary accordingly with the tenure of the project. For example, repeated use of seismic survey at Sleipner field accounts for only a fraction of the net storage cost (Myer et al., 2003). Benson et al., (2005) have estimated the life cycle monitoring cost for storage in EOR fields as well as in saline formations for the basic (periodic seismic surveys, microseismicity, wellhead pressure and injection-rate monitoring) and enhanced monitoring package which include periodic well logging, surface CO_2 flux monitoring and other advanced technologies in addition to basic package. However, utilization of CO_2 as a resource have significant economic potential compared to only storage and treating CO_2 as an industrial waste (Tcvetkov et al., 2019).

8. Conclusions and future prospects

This work presents the past, recent and ongoing developments in CO_2 storage in saline aquifers in favour of reducing CO_2 concentration in the atmosphere and hence mitigate the climate change issue. This review discusses the underlying mechanisms and their optimisation techniques to maximise the security of the injected or stored CO_2 for a longer period. Attention has also been given extensively on each corner of the CO_2 sequestration process such as site characterization,

storage capacity and screening criteria along with existing monitoring techniques and economic viability. In addition, various worldwide CO_2 sequestration projects have been demonstrated. The findings of the present study show that the sequestration of CO_2 in geological saline formations is controlled by various aspects. However, it still requires more research and development to overcome the existing and upcoming challenges. Based on this review, the following conclusions are outlined:

CO_2 trapping mechanisms and optimisation methods

Different mechanisms are responsible for trapping of CO_2 into subsurface saline formations that can be optimised by considering the effect of various parameters. Structural trapping greatly depends on caprock integrity, dome curvature, aquifer depth, brine density, impurities, and the injection rate. The CO_2-brine-rock wettability and the CO_2-brine interfacial tension should be further studied for different rock types at various conditions, and their impact on the capillary sealing needs to be addressed. Furthermore, the rate of CO_2 leakage through the caprock needs to be critically investigated specifically for high pressure-high temperature aquifers. Residual or capillary trapping could achieve maximum immobilization of the CO_2 in a subsurface plume over-time. Optimisation of residual trapping can be achieved by a comprehensive reservoir evaluation and characterization. This includes investigating the CO_2-brine-rock wettability that controls the capillary hysteresis and relative permeabilities of the CO_2-brine system. Additionally, the effects of the pore size, aquifer permeability, brine density and heterogeneity need to be quantified as they directly affect the residual trapping. The factors affecting the wettability alteration during the CO_2 injection process in saline aquifers should be deeply investigated. Similarly, the fluid flow of the wetting layer (phase) needs to be studied at pore-scale to understand the underlying mechanisms that control the residual trapping. Solubility trapping is a function of salinity, pH, temperature, presence of non-condensable impurities, injection strategies (co-injection of CO_2 and brine), and geological structures. Nevertheless, the convective mixing is the main process that enhances and accelerates the solubility trapping in saline aquifers. Coupled geochemical-convective-diffusive model needs to be applied for various geological structures, e.g. fractured and faulted formations. The mineral trapping greatly depends on the ion compositions, pH, temperature, pressure and the available nucleation sites. The effect of chemical reactions, that results in mineral trapping of CO_2, on inducing fractures in the caprock or improving the sequestration process through precipitations should receive more attention in the future research.

Site characterization and storage capacity

Preliminary assessment based on the storage capacity, reservoir and fluid properties, and other site screening criteria at basin-scale is essential to find a suitable site for sequestration. Although there are various successful methods available for storage capacity estimation, considering the effect of mineral trapping can pre-estimate the additional storage capacity due its complexity and poorly understood timescale, therefore requires intensive experimental and modelling studies in the future. Moreover, considering the effect of transient pressure build-up in the saline aquifers during CO_2 injection could be helpful to estimate the additional storage capacity. The excessive pressure build-up may result in creating crack or fracture in the caprock and hence leakage of CO_2 to the environment and also mixing of saline water and ground water. Therefore, optimised injection rate is preferable, and overestimation of storage potential should be avoided by taking a proper safety margin.

Monitoring techniques and economic viability

Implementation of proper monitoring techniques and further managements are essential for early detection of any leakage and spatial migration over time, and finally to verify that the stored CO_2 is isolated and will not adversely affect the host ecosystem. Although saline aquifers have large storage capacity that make them preferable for CO_2 sequestration, they require huge investment for installing new infrastructures and reservoir characterization as against to depleted oil and gas reservoirs. However, the cost may be optimised as fewer number of wells are needed in the of case saline aquifers. This is because they possess high permeability and large pores leading to less pressure dissipation with optimum flow ability. Production of the saline water simultaneously while sequestrating (injecting) CO_2 would make saline aquifer as an attractive storage option and utilization of the produced saline water after required treatments doubles the benefits.

Acknowledgment

The authors gratefully acknowledge the financial support of the Universiti Teknologi PETRONAS under YUTP grant (Grant No.: 015LC0-090).

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